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14. ABSTRACT <p>EUV lithography, using 13.5-nm light, is considered to be the preferred technology for next generation microelectronic circuit fabrication. The introduction of EUV Lithography was delayed already several times and is in danger to be delayed again due to the lack of a high enough power light source allowing for volume production. In this work we investigate whether High Harmonic Generation can be used for such a light source. We build a high energy tunable visible Optical Parametric Amplifier, and drive High Harmonic Generation in Argon and Helium. We study how the efficiency, and the highest producible photon energy, depend on the driver wavelength. We are able to observe EUV light up to the desired wavelength of 13.5 nm. The results agree well with a previously developed theoretical model. We predict that using a 630-nm driver in Helium could have a conversion efficiency of about 10-5. Unfortunately, our pulse energy was too low to confirm this experimentally. This efficiency is too low to construct a EUV source useful for lithography. However, further investigations such as the use of cavity enhancement are necessary to ultimately decide, whether HHG is a viable path to an efficient EUV source.</p>					
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1. INTRODUCTION

The electronics community has always aimed to the miniaturization of integrated circuits (ICs). The technique which is currently used to produce ICs is photolithography, or optical lithography, which consists in shining a photo resist, a material sensitive to light, with an optical beam. The photo resist substrate is the circuit board, and the materials used to produce circuits are chemically deposited on the pattern drawn by light on the photo resist.

Current techniques are capable of producing sub-100-nm features by using UV light at 193 nm from excimer lasers, but for further miniaturization it is necessary to use shorter wavelengths.

Extreme UV (EUV) lithography is one of the most promising techniques within Next Generation Lithography (NGL). EUV lithography uses light at 13.5 nm (92 eV), which is the wavelength at which Mo/Si mirrors have a relatively high 72% reflectivity [1].

The present sources of such light are either discharge or laser produced, and suffer from low efficiency, incoherence and non-directionality.

The High Order Harmonic Generation (HHG) technique allows producing light in the EUV region, starting from a driver pulse in the visible or infrared spectral range. This technique is based on shooting a high intensity ($\sim 10^{14}$ W/cm²) femtosecond laser pulse into a material (usually a noble gas). The driver beam significantly distorts the potential energy seen by the electron in the gas, which can then tunnel through the potential barrier. The electron is then accelerated within a half-cycle of the electric field and upon return to the atom, with high kinetic energy, may recombine with the gas atom. While recombining into the ground state, the difference energy between ground and excited states is emitted through photons in the EUV range. Due to photon energy conservation, the EUV photon energies are high order harmonics of the driver wavelength.

HHG efficiencies, i.e. the conversion of laser energy into single harmonic energy, can reach levels up to 10^{-6} , depending on various conditions (driver energy and intensity, type of gas, gas pressure). Cavity enhancement techniques could boost the efficiency [2] to $\sim 10^{-4}$. These efficiencies are comparable to other EUV-sources, with the added advantage of directionality, coherence, and being free of debris generation, which may contaminate the downstream optics.

Typical HHG spectra are made up of a plateau region, where the energy is rather constant, and a cutoff region where the energy quickly drops. The efficiency (driver to HHG conversion) decreases roughly with the 5th or 6th power of the driver wavelength [3,4], meaning that the HHG process is more efficient with a blue than with a red driver. Conversely, the HHG cutoff energy increases, by theory, with the $1.6^{\text{th}} \div 2^{\text{nd}}$ power of the driver wavelength [3, 5], so a red driver has a higher cutoff energy than a blue driver.

This means that the most efficient driver wavelength to produce photons in a given EUV region is the shortest possible whose cutoff energy is slightly higher than the target (the efficiency starts to drop at the cutoff, so the highest efficiency is at the last harmonic before the cutoff).

Within this project, we have developed a tunable Optical Parametric Amplifier (OPA) in the visible range to study the HHG conversion efficiency and cutoff energy vs. the driver wavelength to identify the optimum drive wavelength for a light source for EUV lithography.

This report is organized as follows: in section 2 we describe the experimental setup, in section 3 we describe and discuss the efficiency scaling experiment, in section 4 we describe and discuss the cutoff energy scaling experiment, in section 5 we drive conclusions and describe perspectives.

2. EXPERIMENTAL SETUP

The experimental setup, shown in Fig. 1, is composed of:

- a commercial Ti:sapphire system;
- a 3-stage visible OPA;
- a HHG setup.

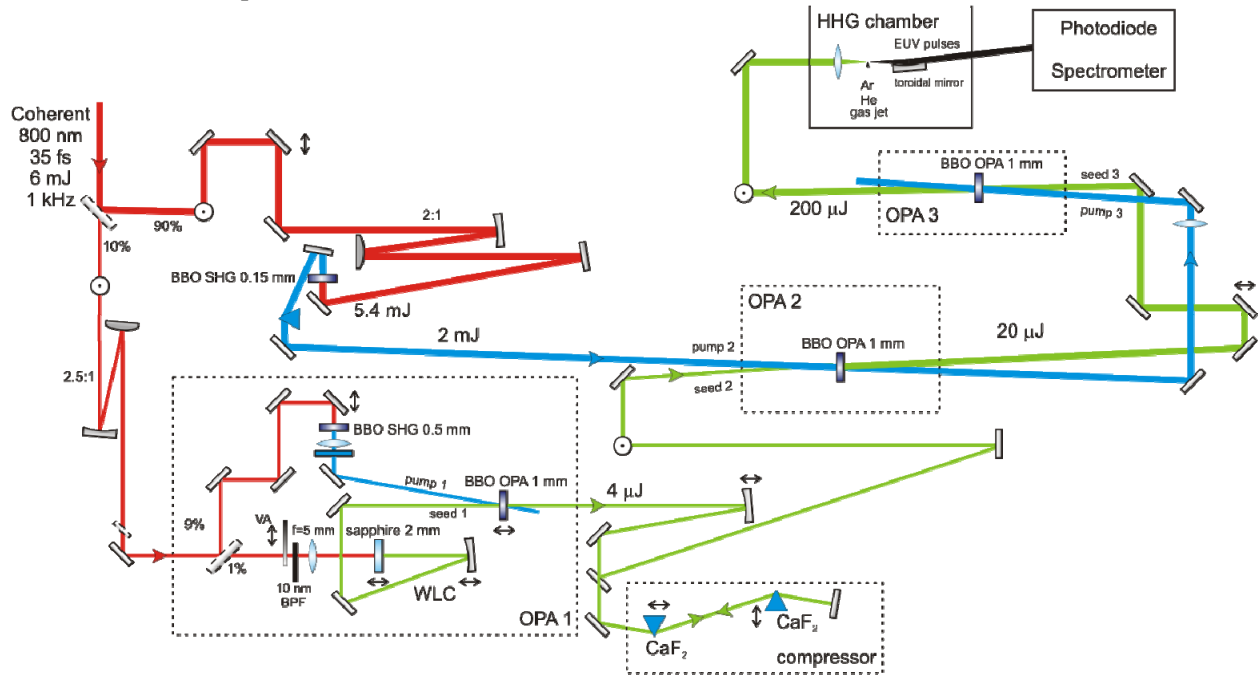


Fig. 1: Experimental setup.

Commercial Ti:Sapphire system

A Menlo-Systems Ti:Sapphire oscillator produces nJ pulses with 30-fs duration at a 85-MHz repetition rate. These pulses are amplified in a Coherent Chirped Pulse Amplification (CPA) system including a grating stretcher, a regenerative amplifier, a single pass amplifier and a grating compressor. The output is a 6-mJ pulse train with 35-fs duration centered at 800 nm with a 1-kHz repetition rate.

3-stage visible OPA

The CPA output pulses are used to pump and seed an OPA in the visible range built according to the scheme demonstrated in [7], with some differences due to the shorter pulse duration of the pump laser (35 vs. 130 fs). This difference poses practical difficulties due to the ease in generating undesired third order effects like Self-Phase Modulation (SPM) and in the higher pulse-to-pulse instabilities.

The CPA output is first split into two uneven parts: 10% of the energy (0.54 mJ) is used for the first stage seed and pump, while the remaining 90% (5.4 mJ) is used to pump the second and third stages.

A 2.5:1 mirror telescope is used to reduce the size of the low energy beam. This beam is further split into 10% (1% of the main beam) used for the seed generation and 90% (9% of the main beam) used for the first-stage pump. The seed is generated by focusing the 1% beam, after a Variable Attenuator (VA) and a 10-nm Band Pass Filter (BPF) at 800 nm, into a 2-mm-long sapphire plate. The process of White Light Continuum (WLC) generation produces low energy pulses in the visible range that are used to seed the first OPA stage. The BPF is used to increase the WLC stability by eliminating the spectral tails in the 800-nm pulses.

The 9% beam is directed through a delay line to a 0.5-mm-thick β -Barium Borate (BBO) crystal cut at 29° for type I phase matching, where Second Harmonic Generation (SHG) occurs. The crystal output produces 40- μ J pulses at 400 nm used to pump the first OPA stage. Pump and seed are focused in a 1-mm, 31° , type I BBO crystal for the first OPA stage, producing a visible output with energy up to 4 μ J. The first stage output is collimated, partially compressed through a CaF_2 prism pair at Brewster angle, and directed to the second OPA stage after a periscope flipping its polarization.

The 90% part of the beam reflected by the first beam splitter is sent to a delay line and to a 0.15-mm, 29° , type I BBO for SHG. The 400-nm output energy is 2 mJ, which are used to pump the second and third OPA stages.

The 2-mJ pump beam amplifies in the second OPA stage the 3- μ J output pulse from the first stage up to 20 μ J in a 1-mm, 31° , type I BBO crystal. The residual pump going through the crystal is used in a third OPA stage to further amplify the visible pulses up to 200 μ J (depending on the wavelength) in a BBO crystal of the same kind.

The rms value of energy fluctuations, normalized over the average energy, is 2.5%, measured over 9 minutes. The M^2 value of the beam is 1.9. The superfluorescence level is below 0.5% of the signal energy.

Fig. 2 (a) shows the tunability of the output spectra. The 4 spectra shown have been used to drive the HHG experiments. The pulse duration was measured with a self diffraction background-free autocorrelation. Fig. 2 (b) shows a sample autocorrelation, in particular for the OPA at 589 nm (duration: 34 fs). The duration varies between 34 and 46 fs depending on the wavelength.

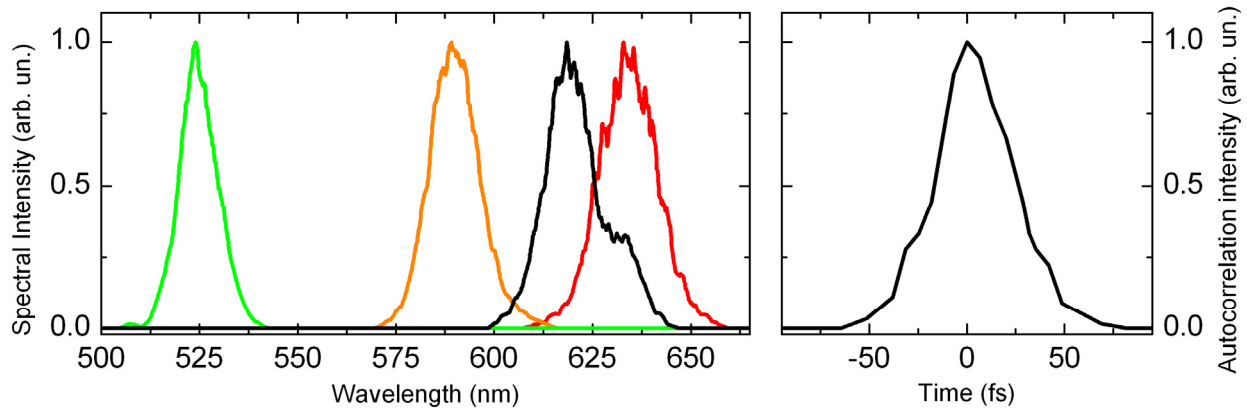


Fig. 2: Spectra and autocorrelation of the second experiment of the visible OPA. The spectra shown were used to drive the HHG experiment. The autocorrelation is relative to the 590 nm pulse and corresponds to 34 fs FWHM pulse duration.

HHG setup

The OPA pulses are sent into a vacuum chamber and focused in an Argon (lens focal length 150 mm) or Helium (focal length 100 mm) gas jet where they drive HHG.

A photodiode and a spectrometer measure the HHG energy and spectrum. We use Al and Be filters in the spectral region before and after 70 eV respectively.

Fig. 3 shows some preliminary spectra in Argon and Helium for the indicated driver wavelengths.

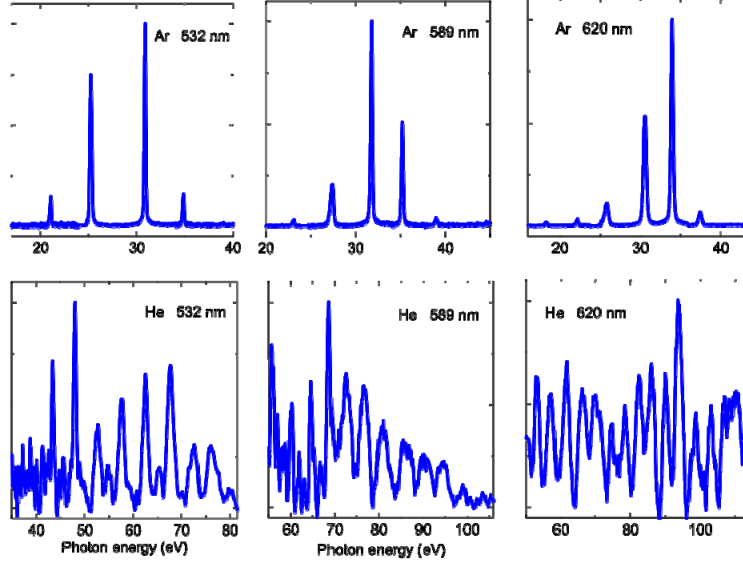


Fig. 3: EUV spectra observed in Argon (first row) and Helium (second row).

3. EFFICIENCY SCALING

We used the OPA to drive HHG in an Argon gas jet, with a 40 mbar pressure (500 mbar backing pressure), and measure the driver to single harmonic order efficiency around 32 eV. We drive HHG with 3 wavelengths generated by the OPA, namely 524 nm, 589 nm and 633 nm, and with the fundamental and second harmonic frequencies of the laser source, i.e. 800 nm and 400 nm. The spectra of the OPA and one of the autocorrelations are shown in Fig. 5. We measure the beam size in the focal position with the knife edge method and keep similar intensities for all 5 driver wavelengths. Table 1 summarizes the characteristics of the 5 driver pulses.

Table 1: Characteristics of the driver pulses used for the HHG efficiency vs. driver wavelength scaling study.

Driver wavelength (nm)	Duration FWHM (fs)	Size at focus ($\mu\text{m} \times \mu\text{m}$)	Energy (μJ)	Intensity ($\times 10^{14} \text{ W/cm}^2$)
400	26	26x26	85	3.1
524	39	33x23	82	1.8
589	34	34x17	85	2.8
633	36	31x18	82	2.6
800	35	21x21	85	3.5

Fig. 6 shows the EUV spectra obtained in the 5 experiments for the indicated driver wavelengths. In the 400-nm case the signal is much higher than in the other cases, so the peak is not shown. The “*” symbol shows the photon count number that has been considered for the wavelength study.

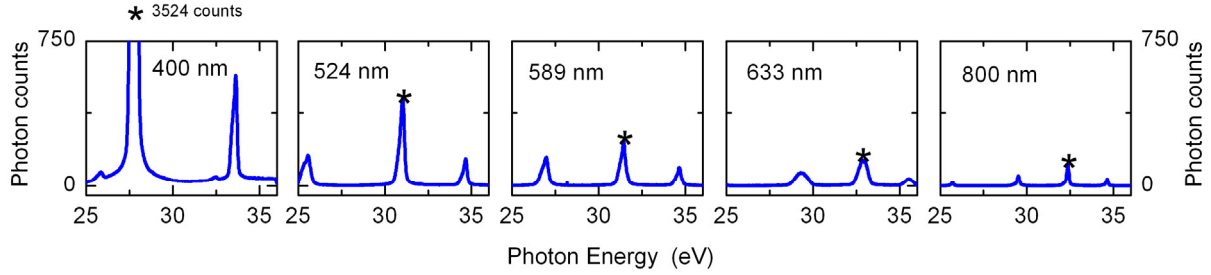


Fig. 6: HHG spectra for the 5 different wavelengths of the efficiency study (linear scale).

Fig. 7 shows the normalized number of photon counts observed in the spectrometer. As expected, the efficiency decreases as the wavelength increases. The result fits very well with the theoretical predictions, presented for example in [3], or [4], where an efficiency scaling of λ^{-5} ÷ λ^{-6} is predicted. While the 3 experimental points observed with the OPA fit very well with the theoretical models, the two points at the fundamental frequency and at the second harmonic of the laser show a slightly higher efficiency. This might be due to the better beam quality in the laser beam than in the OPA, and also to their higher intensity, as indicated in Table 1.

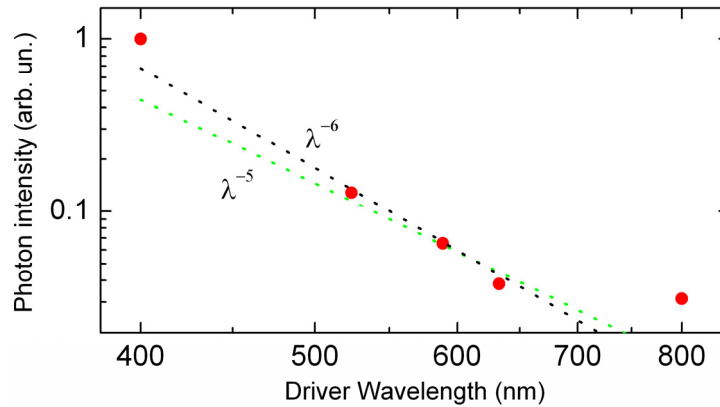


Fig. 7: Efficiency scaling in Argon vs. visible driver wavelength. (Red: experimental data). Black and green dots: theoretical predictions).

4. CUTOFF SCALING

In another experiment we studied the cutoff energy vs. the driver wavelength in Helium (interaction pressure: 50 mbar, backing pressure: 3 bar). Table 2 summarizes the characteristics of the pulses that were used in the experiments. Fig. 8 shows the experimental points and the good agreement with the theory from [3] and from [5].

Table 2: Characteristics of the driver pulses used for the HHG cutoff energy vs. driver wavelength scaling study.

Driver wavelength (nm)	Duration FWHM (fs)	Size at focus ($\mu\text{m} \times \mu\text{m}$)	Energy (μJ)	Intensity ($\times 10^{14} \text{ W/cm}^2$)
400	26	26x26	121	4.4
524	39	23x16	127	5.6
589	34	24x12	119	7.7
618	46	20x13	119	6.3

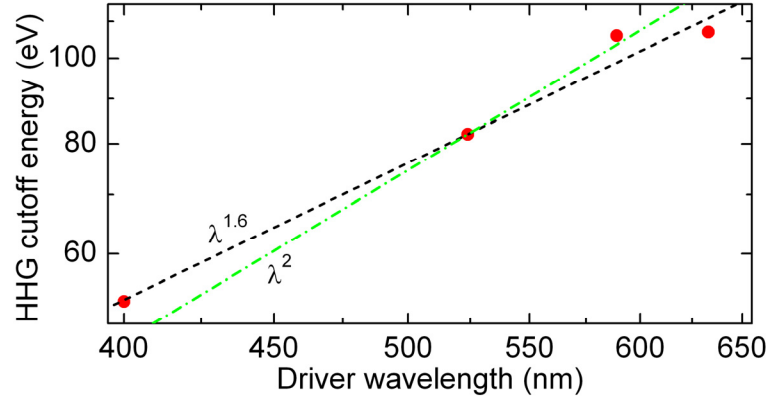


Fig. 8: HHG cutoff energy scaling vs. driver wavelength.

5. CONCLUSIONS AND PERSPECTIVES

In conclusion, we have built a tunable, visible OPA and used it to demonstrate the the scaling of HHG using visible driver pulses, which are ideal for generation of photons at 13.5 nm, the wavelength used in EUV lithography. We have demonstrated cutoff scaling and efficiency scaling, and obtained good agreements to current theories. Further developments may lead to the demonstration of even much higher efficiencies, by energy scaling of the OPA, and usage of cavity enhancement techniques. Eventually, the highest efficiency is achieved by the developing a direct laser source to drive HHG at 13.5 nm.

Based on these results, a promising source that can be developed is the frequency doubled Cr:forsterite, emitting femtosecond pulses at 625 nm. These developments may lead in the future to a source for EUV lithography enabling the mass production of cheap and more compact electronic circuits.

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Publications from this project.

Journal publications

Giovanni Cirmi, Chien-Jen Lai, Eduardo Granados, Shu-Wei Huang, Phillip Keathley, Alexander Sell, Kyung-Han Hong and F. X. Kärtner, "Tunable high energy Optical Parametric Amplifier in the visible for High Harmonic Generation", Optics Express, in preparation.

Giovanni Cirmi, Chien-Jen Lai, Eduardo Granados, Shu-Wei Huang, Phillip Keathley, Alexander Sell, Kyung-Han Hong and F. X. Kärtner, "Cutoff energy scaling of High Harmonic Generation in the 90 eV region", J. Phys. B, in preparation.

Chien-Jen Lai, Giovanni Cirmi, Eduardo Granados, Shu-Wei Huang, Phillip Keathley, Alexander Sell, Kyung-Han Hong and F. X. Kärtner, "Efficiency scaling of High Harmonic Generation in Argon from a visible Optical Parametric Amplifier", Appl. Phys. Lett. in preparation.

Conference publications

Giovanni Cirmi, Chien-Jen Lai, Eduardo Granados, Shu-Wei Huang, Phillip Keathley, Alexander Sell, and F. X. Kärtner, "Efficiency Scaling of High Harmonic Generation driven by a tunable Optical Parametric Amplifier in the Visible", submitted to HILAS 2012, 19 March - 21 March 2012, Laser Optics Berlin, Berlin, Germany.

Chien-Jen Lai, Giovanni Cirmi, Eduardo Granados, Shu-Wei Huang, Phillip Keathley, Alexander Sell, and F. X. Kärtner, "Efficiency and cutoff Scaling of High Harmonic Generation driven by a tunable Optical Parametric Amplifier in the Visible range", in preparation for CLEO 2012.

Giovanni Cirmi, Chien-Jen Lai, Eduardo Granados, Shu-Wei Huang, Phillip Keathley, Alexander Sell, and F. X. Kärtner, "Efficiency and cutoff Scaling of High Harmonic Generation driven by a visible Optical Parametric Amplifier", in preparation for Ultrafast Phenomena 2012.